



INCIDENCE AND SIGNIFICANCE OF POLYNUCLEAR AROMATIC HYDROCARBONS IN THE WATER ENVIRONMENT

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INTRODUCTION

It has been established that many polynuclear aromatic hydrocarbons (which will be referred to as PAH) are carcinogenic to animals and probably to man. This review will focus on their occurrence, significance in, and passage through the water environment, which represents a possible danger to man in terms of widespread exposure to PAH. Literature was reviewed for the years 1968 through the first half of 1973 as an updating of an earlier comprehensive review by Andelman and Suess,¹ which covered the literature prior to 1968, and to which the reader should refer for additional information.

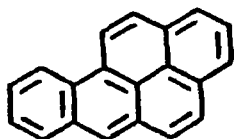
Most of the studies on the incidence of PAH in the water environment have been carried out in Europe. The most comprehensive investigations on the sources of PAH, their spread in the environment, and the efficacy of various treatment methods have been done by Borneff and his associates in the German Federal Republic. Soviet investigations, principally by Il'nitskii and Shabad, have concentrated on effluents from various manufacturers as one of the main sources of pollution of water bodies by PAH.

In a recent review article, Shabad and Il'nitskii² pointed out that, although the amount of PAH a man may consume from heavily polluted water is only about 0.1% of the amount he consumes in food, carcinogenic PAH from water can accumulate in various aquatic organisms, including fish, some mollusks, and edible algae. Water used for irrigation may also affect the level of carcinogenic PAH in vegetable foodstuffs.

The need to establish hygienic standards for carcinogenic compounds has been given more attention in the last few years. Shabad³ states that "without waiting for the complete elucidation of mechanisms of blastomogenesis or even the solution of the problem of the action threshold of carcinogens, everything should now be done to establish maximum permissible doses and concentrations of individual carcinogens, taking account of the nature and duration of contact with them. The PAH are the first priority." Similarly, the most recent edition of the World Health Organization European Standards for Drinking Water (1970)⁴ recommends that "for the safety of consumers, the concentration of PAH should not exceed 0.2 $\mu\text{g/l}$." It further recommends that there should be at least one center in each country

able of carrying out investigations of PAH in drinking water, and that more research into their presence and importance in drinking water is required. The most recent (1962) United States Public Health Service standards for drinking water do not mention PAH. However, a recent publication of The National Institute of Environmental Health Task Force³ states: "Some materials which enter waterways are known to be carcinogenic to animals. Others may have physiological activity of which we are currently unaware. The organic contaminants should be identified and the physiological significance of the concentrations found in the environment determined. Also the physiological significance of the heterogeneous mixtures as they exist in water should be evaluated."

Typical polynuclear aromatic hydrocarbons



3,4-benzpyrene

3,4-Benzpyrene (BP) is ubiquitous in the environment and one of the most potent of the carcinogenic PAH. Consequently, it has been the most studied. 1,2,5,6-Dibenzanthracene (DBA) has also been identified as a very potent carcinogen and 1,2-benzanthracene (BA), 3,4-benzfluoranthene (3,4-BF), 10,11-benzfluoranthene (10,11-BF), chrysene (CH), 9,10-dimethyl-1,2-benzanthracene (DMBA), indeno (1,2,3-cd) pyrene (IP), 3-methylcholanthrene (MCA), and 1,2-benzpyrene (1,2-BP) have been shown to possess carcinogenic properties.¹

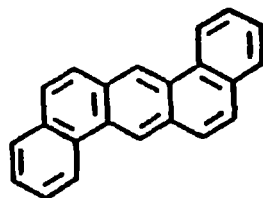
The two most important properties influencing the incidence of PAH in water are their solubility and stability. Their high molecular weight and lack of polar substituent groups make them very insoluble in water. The solubility of BP in clean river water or tap water was found to be approximately 0.01 µg/l.⁴

Despite the low solubility of PAH, their presence in environmental waters can be increased by the action of micelles formed by synthetic detergents and by the action of other soluble

The following review will deal with the pertinent physicochemical properties of PAH, their analysis, origin, source, and transport in the environment; their presence in environmental waters, their removal or destruction by various treatment processes, and the possible health consequences to man of PAH in the water environment. The emphasis will be on 3,4-benzpyrene, one of the most carcinogenic of the PAH.

PHYSICOCHEMICAL PROPERTIES AND ANALYSIS OF PAH

The chemical structures of two typical PAH are as follows:



1,2,5,6-dibenzanthracene

organic compounds. The presence of surfactants in the range of 10 to 50 mg/l increases the solubility of BP from two to ten times.⁷ However, as noted by Andelman and Suess,¹ the concentration of surfactants in natural and treated waters rarely reaches a high enough concentration to affect the solubility of BP. Surface active agents in the concentrations from 0.1 to 1.0 mg/l do not show any effect on BP solubility.⁴ Drinking water standards typically permit only 1.0 mg/l of surface active agents.

Phenol concentrations from 1 to 50 mg/l do not have any effect on BP solubility,⁸ but the addition of caffeine to water increases both the solubility and rate of BP going into solution, as the caffeine concentration is increased from 0.01 to 0.10 mol/l.⁹ In their review, Andelman and Suess¹ noted that other studies showed that lactic acid, acetone, and ethyl alcohol increase the solubility of BP in water. The possibility exists that other organic compounds present in polluted waters could also solubilize BP. Finally, highly polluted water containing emulsions of organic solvents

would be capable of containing relatively large amounts of BP.¹

Studies have also shown that BP can be sorbed and concentrated on activated carbon, calcareous material, silica, glass, and soil particles. The presence of these minerals and other suspended and settled particles in natural waters can also be sources of relatively high concentrations of BP.¹

BP can be very stable in water and remain in solution over long periods of time. Il'nikskii et al.⁷ performed experiments using pond and tap water with initial BP concentrations of 10 and 0.01 µg/l. The 10 µg/l solution is prepared by adding 1 ml of acetone solution containing 2 µg of BP to a 250 ml flask, evaporating the acetone, adding 200 ml of tap or pond water, and mixing thoroughly. The 0.01 µg/l solution is prepared by adding 10 ml of acetone containing 0.01 µg/ml BP into a 10 l jar, filling the jar with pond or tap water, and mixing thoroughly. Both solutions are stored in the dark. After extracting the solutions with benzene, quantitative analysis is performed spectrofluorimetrically using Shpol'skii's effect. Several experiments showed that undecomposed BP could still be detected after 35 to 40 days to the extent of 5 to 20% of the initial concentration. At the concentration of 10 µg/l BP, only 10 to 15% of the BP was destroyed in the first few days. However, they found that at an initial concentration of 0.01 µg/l, almost 50% of the BP was destroyed in 24 hr. They also found that the decomposition took place more rapidly in 0.02 N potassium permanganate than in pure tap water, and in acid solution (pH 2.7) faster than in alkaline (pH 11.6) and neutral solutions (pH 6.7).

In other experiments, photooxidation was found to destroy 56% of the BP (present at an initial concentration of 0.1 g/l) in one day. In the dark, the degradation was negligible. With the addition of sodium sulfite, the decomposition of BP was increased to 75% in both the presence and absence of light.⁹

In general, studies have shown that the higher the temperature, oxygen concentration, and light intensity, the faster the rate of BP decomposition.¹

The greatest problems encountered in the analysis of PAH are their low concentrations and the separation of PAH from other organics as well as from each other. Preliminary techniques of extraction and concentration with solvents such as benzene, pentane, n-heptane, n-hexane, and n-

octane are necessary. These preliminary techniques can be sources of error because even the purest solvents contain traces of PAH. Fedonin et al.¹⁰ noted that these traces may affect the ultimate analytical results because these preparatory techniques of separation and concentration involve the use of large amounts of solvents which, when evaporated, could contain considerable amounts of PAH. It is, therefore, imperative that a blank correction be made in procedures using large amounts of even chemically pure solvents.

Column, liquid, paper, and thin-layer chromatography are the most frequently used methods for the separation of PAH, followed by quantitative analysis using ultraviolet-visible absorption spectra and fluorescence spectra. Several studies that demonstrate the methodology for the examination of a wide variety of water and wastewater samples will be mentioned in the following paragraphs.

Paper chromatography of a mixture of PAH has been discussed, in which the minimum detectable amount on a chromatogram was 0.026 µg BP, 0.022 µg 20-methyl-cholanthrene, and 0.062 µg BA.¹¹ Schloz and Altmann¹² detected 10⁻⁹ to 10⁻¹³ g/ml of BP in water by fluorescence spectroscopy, after a tenfold enrichment by extraction with cyclohexane, a 100-fold enrichment by evaporation of the extract, and a subsequent separation by thin-layer chromatography.

Borneff¹³ described a method to be used routinely for the examination of PAH in drinking water and ground water to assess the efficiency of a surface water purification plant. The World Health Organization⁴ recommends this procedure for the determination of six PAH in drinking water to assess the presence of carcinogenic PAH in drinking water. Another method is described by Jaeger and Kassowitzova¹⁴ that can detect BP in drinking water at a concentration as low as 0.003 µg/l. Siddiqui and Wagner¹⁵ describe a method for the determination of BP and 3,4-BF in rain water, ground water, and wheat using extraction with benzene, saponification with 2 N KOH, separation by either column or thin-layer chromatography, detection with ultraviolet light, and photometric determination.

The determination of PAH in polluted surface waters can be particularly difficult because of the presence of large quantities of organic compounds and the very low concentrations of PAH. Samples from the Charles River Basin (Boston, Massachusetts) were analyzed for PAH and other

ic compounds using computerized gas chromatography, mass spectrometry, and high pressure liquid chromatography.¹⁶ Keegan¹⁷ describes a method developed for the determination of PAH in parts per trillion range and utilized for all rivers in Connecticut.

Industrial effluents are one of the major sources of PAH contamination of surface waters. Ershova and Mints¹⁸ detected and quantitatively determined BA in petrochemical effluents by a variant of their spectroscopic-fluorescence method. A method is described for the determination of a mixture of several PAH in wastewaters using thin layer chromatography on alumina and low temperature spectral luminescent analysis.¹⁹ Cardoni and Thiella²⁰ used gas chromatography to do simultaneous qualitative and quantitative analysis of aromatic hydrocarbons, including PAH and phenols in industrial wastewaters.

ORIGIN, SOURCE, AND VEHICLES OF TRANSMISSION OF PAH

For the purposes of the following discussion, the term "origin" will refer to the formation of PAH, and the term "source" will refer to the medium or material in which PAH can concentrate from which they can be disseminated to the environment. Industrial effluents, domestic effluents, precipitation, atmospheric fallout, and surface water are the vehicles of transmission of PAH.

Origin and Source of PAH

Despite their low solubility and stability in water, PAH are nevertheless ubiquitous in the environment. To understand this, it is necessary to determine the origin of PAH.

Industrial operations engaged in the pyrolytic processing of organic raw materials, such as coal and petroleum, form PAH when the process is carried out at high temperatures (e.g., 700°C), as compared to the formation of paraffins, cycloparaffins, olefins, and phenols at lower temperatures (0 to 450°C). The consumption of these pyrolytic products (coal tar, coal tar pitch, creosote) can also produce PAH.

Some of the industrial operations that produce PAH are the preparation of acetylene from natural gas,^{21,22} the pyrolysis of kerosene to benzene, toluene, and other organic solvents,²³ the pyrolysis of wood,²⁴ manufacture of electrolytic al-

uminum using graphite electrodes,²⁵ coke production,²⁶ gas production from petroleum,²⁶ the production of synthetic alcohol,²⁶ and oil refinery operations.²⁷⁻²⁹

The combustion of fuels by automobiles, airplanes, and boats involves the production of PAH. The source of PAH in the Charles River (Boston, Massachusetts) was thought to be automobile exhaust condensate washed from the streets directly into the river by rainfall.¹⁶ Studies of soil, snow, and vegetation samples taken in the vicinity of a Moscow airport have been shown to contain large quantities of PAH.³¹ Significantly larger amounts of BP were found in a Moscow reservoir with high boat traffic than a similar reservoir with low boat traffic.³² Carbon black, used in the manufacturing of automobile tires, contains PAH and consequently the wear of tires on the road can be sources of PAH. Asphalt used in road construction contains PAH that could also be a source of contamination.¹

The presence of PAH in remote, nonindustrialized areas raises the question of the endogenous formation of PAH by plants and microorganisms. Borneff et al.,³³ using the technique of labeled carbon assimilation, conclusively demonstrated that PAH can be synthesized by plants. Bacteria and phytoplankton are also capable of producing PAH. The appearance of BP in in vitro cultures of three strains of marine phytoplankton was corollary to the appearance of a mixture of aerobic bacteria. The addition of antibiotics to inhibit bacterial growth also prevented the formation of BP.³⁴ Forest soils sterilized at 120°C and 170°C and seeded with either *Clostridium putride* or *Escherichia coli* contained significantly larger amounts of BP after standing at room temperature for six months.³⁵ The possibility that bacteria are capable of synthesizing BP is supported by the fact that compounds similar to BP have been found in the intestinal region of many animals where various types of bacteria are also found. Hydrocarbons of the BP type are found in all organs, particularly the liver, kidneys, spleen, and lungs.³⁶

The Clipperton Atoll Lagoon in the northeast Pacific Ocean is not located near any source of BP, yet its waters were shown to contain 3.5 to 4.0 µg/l of BP.³⁷ However, its water is rich in phytoplankton and marine bacteria, which seems to confirm the possibility of the biosynthesis of BP in a marine environment.³⁸

Finally, it should be noted that, although no

PAH have been isolated from uncracked crude oil, the possibility exists that they contain traces that could be sources of PAH to environmental waters. The fact that large quantities of such oil are present in marine waters (it is estimated that one million metric tons are lost in the ocean during transport alone) and can accumulate in various marine organisms suggests that more research be done on this potential source of PAH.^{39,40}

Vehicles of Transmission of PAH

Industrial effluents carry PAH into receiving waters and existing methods of treatment generally do not reduce their PAH content. Effluents from industries engaged in the thermal processing of mineral fuels are one of the largest sources of PAH contamination of surface waters. Most of the quantitative studies of PAH in industrial effluents have been done by Soviet authors. Ershova and Mints^{18,26} found BP and BA in effluents from industries engaged in the production of gas from petroleum and in the production of synthetic

alcohol. Samoilovich and Red'kin²⁸ investigated effluents from oil refineries and concluded that they were the source of BP pollution of the Sumzha River. Ershova²⁹ detected BP in effluents from several oil refineries along the Volga River. River samples taken below the refineries and a sand sample from the filter of a waterworks located downstream all contained BP. The results point to the refinery effluents as the sources of the BP contamination. The amounts of PAH found in typical effluents are summarized in Table 1.

Since municipal wastes often contain large amounts of industrial effluents, they can transport PAH into surface waters. Run-off water passing over roads, plants, and soils carries PAH into the sewage system. In addition, human urine has been shown to contain BP.¹ Table 2 lists some concentrations of PAH found in domestic wastes.

The amount of BP emitted annually into the atmosphere of a large industrial city has been estimated to be 30 to 40 kg.²⁹ Substances polluting the air are eventually deposited by

TABLE I
PAH Concentration in Industrial Effluents^a

Industry	Source of waste water	BP concentration (µg/l)
Shale-oil	After treatment for dephenolization	2-320
Coke by-products	Not indicated	Present
	After biochemical treatment	12-16
	After oil separation (5 samples)	6.5, 130, 250, 290, and "big" quantity
	Spent gas liquor	Very small quantity
Coke or oil-gas works	Before discharge to sewer	Not indicated* 1,000 and 340*
	Before discharge to sewer (2 plants)	
Oil-gas works	After oil separation (3 samples)	3, 6, and 30
Oil refinery	After oil separation (3 samples)	None detected
Tar paper	Not indicated	Present
Acetylene	Not indicated	15-100
Ammonium sulfate	After cooling and settling	About 10*

*Other PAH also present

*Taken from Andelman and Suez.¹

TABLE 2
PAH Concentration in Domestic Effluents^a

Source	Concentration (µg/l)		
	BP	Carcinogenic PAH	Total PAH
Domestic effluent from a small community after primary treatment	0.170	3.0	15.0
Waste effluents entering Rotach River:			
Sample 1	0.015	0.2	0.5
Sample 2	0.047	1.1	2.7
Sample 3	0.079	1.0	6.6
Sample 4	0.100	0.5	5.1
Sample 5	0.038	0.3	0.8
Waste effluent of Stockach	0.100	5.0	15.0
Waste effluent of Radolfzell	0.368	2.8	8.6
Waste effluent of Hegne:			
Sample 1	0.001	0.1	0.8
Sample 2	0.011	1.2	4.2
Sample 3	1.840	37.9	87.5
Sample 4	0.450	31.6	68.0
Vegetable cold wash water:			
Sample 1	1.60	8.1	25.2
Sample 2	1.02	5.8	22.6
Urine from Paris inhabitants (4 samples)	1-3	-	-
Sewage of Leningrad	Present	-	-
Sewage, final effluent (4 plants-8 samples)	Present	0.15-1.50	-
Sludge from secondary treatment (humus)	Present	Present	-
Humus, dried	3,000	-	-

^aTaken from Andelman and Sues.

natural precipitation or fallout directly into surface waters or onto plants and soil where they can be carried by run-off into surface waters. Due to the endogenous formation of PAH, a "background" level of PAH is present in most soils. After an extensive study of soil samples in remote areas of Russia, Shabad et al.²⁹ concluded that BP concentrations of 5 to 10 µg/kg of soil are not unnatural levels associated with exogenous contamination,

while concentrations of 1 to 2 µg/kg are typically background levels. The same authors found as much as 220,000 µg of BP/kg of soil in the vicinity of an oil refinery. Shcherbak²³ found that vegetation samples taken within 300 m of the "neftgaz" plant in Moscow contained BP in concentrations varying from 0.6 to 5.9 µg/kg of dried material, and that most soil samples taken 3,000 m from the plant still contained quantities of BP higher than

the general background level for the city of Moscow. Studies have also shown that PAH can be concentrated in deeper soil layers and hence also contaminate the ground water.²³

PAH have also been found in snow samples taken in the vicinity of a Moscow airport,³¹ on the grounds of a timber-chemical plant,³⁴ and in various areas of a large industrial city (Moscow).⁴¹ Siddiqui and Wagner¹⁵ also found levels as high as 0.3 µg/l of BP and 3.1 µg/l of 3,4-BP in rain water.

PAH IN ENVIRONMENTAL WATERS

For the purpose of this review, environmental waters will be divided into four different categories: 1) marine waters, 2) surface waters, including flora, fauna, and sediments, 3) ground water, and 4) drinking water.

The most extensive studies of PAH in the marine environment have been done by Mallet and his co-workers. They made systematic studies of the Atlantic, Channel, and Mediterranean coasts of France, the Bay of Naples, Italy, and the western coast of Greenland. BP was found to be present in almost all samples of plankton, algae, fauna, and sediments in the areas studied. Even the remote areas off the coast of Greenland contained on the average the same concentrations of BP as areas along the French coast, thus indicating the ubiquity of BP in the marine environment. The results of Mallet's studies are summarized by Andelman and Sues.¹

As noted previously, large amounts of crude oil are lost to the ocean each year. The possibility that marine organisms can concentrate and fix hydrocarbons could account for their presence in some marine organisms. Lee et al.⁴² analyzed several fish, including anchovies and smelt, from the oil polluted San Diego Bay and found BP in amounts as high as 10 µg per fish, as well as lesser amounts of other PAH (average dry weight of each fish varied from 2 to 10 g). Lee et al.⁴³ also observed that the marine mussel *Mytilus edulis* rapidly took up BP from seawater solutions and retained significant amounts even after being placed in clean seawater. Blumer,⁴⁶ while studying the long-term effects of an oil spill in Buzzards Bay, Massachusetts, found toxic aromatic hydrocarbons still present in edible shellfish 2 months after the oil spill. Fish (*Carassius auratus*) feeding on food containing BP were found to metabolize

and eliminate BP very slowly with residues left in the liver, gonads, and muscles of the fish more than 2 days after the administration of BP was halted.⁴⁴

The presence of BP in the tidal mud of Saint Malo Bay was investigated by Mallet et al.⁴⁵ and found to vary seasonally in BP content due to the action of anaerobic bacteria which synthesize BP and the aerobic bacteria which degrade BP. The presence of BP in the waters of Clipperton Atoll Lagoon was also thought to be due to the combined action of anaerobic and aerobic bacteria.³⁷ BP found in Atlantic phytoplankton in the Greenland to Gascony Gulf areas was thought to be synthesized from the lipids of plankton by bacteria.⁴⁶ The source of most of the PAH in the marine environment is probably due to endogenous sources rather than to exogenous sources, such as surface waters from highly polluted areas.

Other studies have revealed the presence of BP in wide zones of the Adriatic but only during the estival season.^{47,48} Studies conducted by Greffard and Meury⁴⁹ in the Toulon Harbor showed PAH concentrations varying between 9.0 and 284 µg/100 g of dried slime, and BP concentrations varying from 3.0 to 4.0 µg/100 g dry weight in washed mussels.

Considerable quantities of PAH can be carried into surface waters by domestic sewage, industrial effluents, and storm water run-off from roads and contaminated land. The maximum amount determined in any of these sources has been 134 µg/l.⁵⁰ The presence of PAH in areas remote from any exogenous sources also confirms a natural level of PAH in surface waters. Il'nikskii et al.⁵¹ concluded that this level did not exceed 10⁻⁵ to 10⁻⁴ µg/l of BP for surface waters, 1 to 2 µg/kg of BP for bottom sand, and 5 µg of BP per kilogram of dry matter for algae and higher plants. Levels in excess of these amounts can be taken as indications of environmental pollution by man.

Highly polluted waters are conducive to the preservation of PAH in surface waters. The Sunzha River in the U.S.S.R. is heavily polluted with detergents, benzene, ether, and oils, their concentrations ranging from .96 to 350 mg/l. As would be expected, investigations carried out by Samoilovich and Red'kin⁵² showed high BP concentrations at great distances downstream from industrial outfalls (0.07 to 1.06 µg/l at a point 25 km downstream from the lowest outfall).

Laboratory experiments conducted by Sues,⁵³

under conditions similar to the Ohio River (Pittsburgh, Pennsylvania), showed that BP degradation in natural waters depends on solar illumination, temperature, and dissolved oxygen concentrations. In polluted waters with high turbidity, BP would degrade very little in suspended solids and probably not at all in bottom sediments. BP degradation in surface waters is also related to flow velocity and surface area. Samoilovich and Red'kin²⁸ found that the higher the flow velocity and the greater the surface area, the faster the degradation of BP.

PAH concentrations in the surface water environment are very high, essentially due to exogenous pollution. Typical concentration ranges are summarized in Table 3 and the results of some recent studies are summarized in Tables 4 and 5.

Uncontaminated ground waters show the lowest concentrations of PAH of all natural waters. Concentrations will generally not exceed 0.05 µg/l.¹³ Siddiqi and Wagner¹⁵ reported finding 0.01 to 0.03 µg/l of BP and 0.08 to 0.6 µg/l of 3,4-BP in ground water samples.

Borneff and Kunte¹³ noted that concentrations up to 0.10 µg/l of PAH are found in drinking water. They concluded that values between 0.15 and 0.20 µg/l demand an examination of the water treatment facilities, and water containing PAH in excess of 0.20 µg/l should be rejected for human consumption. The World Health Organization European Standards for Drinking Water,⁴ adopted in 1970, also states that concentrations of PAH in

excess of this amount are not safe for human consumption. Studies summarized by Andelman and Sues¹ show that, in general, the total carcinogenic PAH in drinking water do not exceed 0.025 µg/l.

EFFECT OF WATER AND WASTEWATER TREATMENT ON PAH

Since considerable portions of the PAH present in untreated waters are associated with particles, separation processes such as sedimentation and filtration with sand or activated carbon can be very effective in reducing PAH concentrations. The highest removal (98.5%), reported by Borneff,¹⁴ was accomplished in a flocculation plant, which used as a primary step ferrous sulfate and chlorine, followed by ozone and ferric chloride, and then sand filtration. The purified water had a PAH content similar to ground water. Reichert et al.²⁹ found that primary and secondary sedimentation of sewage reduced the PAH by 80 to 90%. Further treatment with synthetic flocculants, followed by filtration through activated carbon, yields a water with PAH concentration similar to ground water. Lawrenz¹⁶ found that treatment of water from the Elbe River in flocculation tanks was effective in reducing PAH concentrations.

Mechanical separation processes will have very little effect on dissolved PAH. Il'nikskii¹⁷ conducted experiments using BP concentrations in the

TABLE 3

Typical Concentration Ranges of BP and PAH in Various Fresh Surface Waters

Source	Concentration (µg/l)			Reference
	BP	Carcinogenic PAH	Total PAH	
Rhine River at various points	0.05-0.11	0.01-0.73	0.73-1.50	Andelman and Sues ¹
Various German rivers	0.001-0.04	0.04-1.30	0.12-3.1	Andelman and Sues ¹
One American river	0.078-0.150	-	-	Keegan ¹³
Rivers receiving effluents from industries that are sources of PAH	0.0001-12	-	-	Andelman and Sues, ¹ Ershova, ²⁸ and Samoilovich and Red'kin ²⁸

TABLE 4
Concentration of BP in Surface Waters

Source	BP concentration µg/l	Reference
Moscow reservoirs, slightly polluted	4-13	Il'nikskii and Rozhkova ¹⁷
Volga River below discharge site of oil refinery	0.0001	Ershova ²⁸
Pskov region, remote from exogenous sources of BP	10 ⁻⁵ -10 ⁻⁶	Il'nikskii et al. ¹⁷
Sunzha River, 3-4 km downstream from discharge sites of petroleum refinery (23 samples)	0.05-3.5	Samoilovich and Red'kin ²⁸
Sunzha River, 25 km downstream from lowest discharge site of petroleum refinery	0.07-1.06	Samoilovich and Red'kin ²⁸
Oyster River, Conn., U.S.		Keegan ¹³
Sample 1	0.078	-
Sample 2	0.125	-
Sample 3	0.150	-

TABLE 5
Concentration of BP in Surface Water Environment

Source	BP concentration µg/kg dry material	Reference
Rublevskoye Reservoir		Il'nikskii et al. ¹⁷
Plankton	0.7-1.8	-
Bottom sand	44	-
Pond weeds	0.6-2.7	-
Khimkinskoe Reservoir		Il'nikskii et al. ¹⁷
Plankton	7.3-8.8	-
Bottom sand	390-500	-
Pond weeds	1.7-37.8	-
Bottom sediments of Sunzha River, 3-4 km downstream from discharge site of refinery	9.2-19.0	Samoilovich and Red'kin ²⁸
Bottom sediments of Sunzha River 25 km downstream from discharge site of refinery	trace-3.1	Samoilovich and Red'kin ²⁸
Silt in Moscow Reservoir	1.8-9.0	Il'nikskii and Rozhkova ¹⁷
Pskov region, remote from exogenous sources of BP		Ershova ²⁸
Algae	5	-
Bottom sand	1-2	-

TABLE 6

Efficiency of Various Combinations of Water Treatment Processes to Eliminate Low Concentrations of BP^a

Process	Number of experiments	Initial BP concentration, $\times 10^{-3}$ g/ml	Efficiency, %
Standing (precipitation)	6	1.7	54 \pm 16
Flocculation + standing	4	3.3	56 \pm 9
Flocculation + filtration in pilot-plant installation	3	1.6	69
	7	1.8	89 \pm 3

^aTaken from Il'nitskii.¹⁷

age that they are usually found under natural conditions to test the efficacy of mechanical separation processes. He found that the techniques investigated reached their practical limit at low concentrations since the techniques were designed to remove suspended, not dissolved, matter. This explains results summarized in Table 6 which show that the same effectiveness for simple standing and of flocculation followed by standing.

Biological treatment is very ineffective in removing any PAH from contaminated waters. A laboratory study, conducted by Malaney et al.,¹⁸ showed that activated sludge treatment of domestic sewage from Nashville, Tennessee, was unable to effect any significant removal of PAH by the oxidative mechanism within normal detention times. Any reduction of PAH in the sewage was due to adsorption by the activated sludge. Ershova¹⁶ found that effluents from a petrochemical plant still contained up to 1.7 μ g/l of BP and 0.6 μ g/l of BA even after being subjected to petrochemical purification in aeration tanks. Il'nitskii et al.¹⁷ also found that biochemical treatment, steam dephenolization, and extraction with butylacetone do not achieve the complete inactivation of PAH in effluents.

The contamination of surface and ground waters with PAH and compounds that may solubilize PAH makes it necessary to investigate methods for the elimination of dissolved PAH from potable waters. Since mechanical methods are not able to remove dissolved PAH, oxidation is especially important. Studies on the efficacy of ozonation, UV and gamma irradiation, chlorination, and treatment with ClO_2 have been carried

out by Il'nitskii, Ershova, Shabad, Reichert, Gabovich, and their associates (References 7, 27, 57, and 59 to 66). Their results indicate that ozonation is the most effective treatment for the reduction of PAH in water, followed by treatment with ClO_2 . The other processes studied were for the most part ineffective in significantly reducing PAH concentrations using normal rates of application and detention times. The efficacy of ozone may be reduced by the presence of other organic compounds. BP was found to be the most resistant of the PAH to treatment. Some test results are summarized in Tables 7 and 8.

Since BP can be present in water in either a dissolved state or sorbed onto particles, Il'nitskii et al.¹⁷ carried out experiments using BP sorbed onto soil particles. The results indicate that the BP sorbed onto soil particles are more stable than dissolved BP. One minute's contact with ozone inactivated 61% of the dissolved BP but only 33% of the sorbed BP; 2½ min contact destroyed all the dissolved BP but only 60% of the sorbed BP.

In order to deliver drinking water to consumers with the lowest possible concentration of PAH, Borneff³⁴ recommends the use of flocculation, ozonation, filtration, and adsorption on activated charcoal for raw waters containing 0.1 to 1.0 μ g/l of PAH; and that raw waters containing over 1.0 μ g/l of PAH never be used even after purification. Il'nitskii¹⁷ concluded that by using ozone rather than chlorine and UV irradiation, a 100-fold greater reduction of BP could be accomplished. For dissolved BP and other PAH, ozonation is necessary to deliver water practically free of carcinogenic hydrocarbons to consumers.

TABLE 7

Concentration of Hydrocarbons After One Minute of Contact with Ozone^a

Hydrocarbon	Mean Residual, %
3,4-Benzopyrene (BP)	39
Pyrene (PY)	15
1,2-Benzanthracene (BA)	4.5
1,2,3,6-Dibenzanthracene (DBA)	3.6
9,10-Dimethyl-1,2-Benzanthracene (DMBA)	0

^aTaken from Il'nitskii et al.¹⁷

TABLE 8

Comparative Stabilities of Certain Polycyclic Aromatic Hydrocarbons to UV Radiation and Ozone^a

Hydrocarbon	Concentration of undecomposed hydrocarbon (%) after	
	UV irradiation for 60 min	Ozonization for 1 min
3,4-Benzopyrene (BP)	95	39
Pyrene (PY)	83	15
1,2,3,6-Dibenzanthracene (DBA)	83	3.6
Pyrene	81	—
1,2-Benzanthracene (BA)	68	4.5
1,12-Benzperylene	45	—
9,10-Dimethyl-1,2-benzanthracene (DMBA)	11	0

^aTaken from Il'nitskii et al.¹⁷

Waters distributed to consumers are usually used for the preparation of food. Studies carried out by Il'nitskii et al.¹⁷ showed that the boiling of water containing 0.0002 to 0.002 μ g/l of BP for 60 min destroyed most of the carcinogen. The results of this experiment are shown in Table 9.

HEALTH CONSIDERATIONS OF PAH IN THE WATER ENVIRONMENT

There is no epidemiological evidence to prove that PAH in drinking water are related to the incidence of cancer. Also, as stated by Il'nitskii and Shabad,¹⁷ the amount of carcinogenic PAH consumed by man in water is typically only 0.1% of the amount he consumes from food. Nevertheless, accumulation of PAH in edible aquatic organisms can greatly increase this amount.^{1,2,4-6}

TABLE 9

Effect of 60 Minutes' Boiling on BP Dissolved in Water at Various pH Levels^a

pH of water	Number of experiments	BP concentration, μ g/l	
		Initial	After boiling
2.7	4	0.001	0, 0, 0, 0.0002
3.0	3	0.0002	0, 0, 0
6.3	4	0.002	0, 0, 0.0002, 0.0002
11.0	4	0.002	0, 0, 0.00095, 0.0002

^aTaken from Il'nitskii et al.¹⁷

The use of contaminated water for irrigation can also spread PAH into other foodstuffs.³

Shabad³ urges that everything possible be done to establish maximum permissible doses (MPD) and maximum permissible concentrations (MPC)

for individual carcinogens, with first priority being given to the PAH, most notably BP. He reported on one such effort based on the intragastric administration of a gradually diminishing series of doses of BP and observing whether or not cancer was induced. These experiments were used to establish doses which did not produce cancer. On the basis of these data and taking into account the differences in body weight and organ surface areas of rats and humans, they calculated that the MPC for BP in water should not exceed 0.0003 µg/l, which for all practical purposes is zero. Even though this type of approach may be controversial and far from perfect, Shabad emphasizes the need for more experiments in this direction, including the transplacental influence on succeeding generations.

Andelman and Suess¹ discussed another concept of MPC in water in which it was suggested that the amount of carcinogens taken up by man in drinking water should not exceed 1/10 of the material taken up from normal urban air. Using this concept the MPC for water would be 0.017 µg/l. Based on an average daily consumption of 2.5 l of drinking water, this comes out to 15 µg/year and about 1 mg in a lifetime. The 1970 World Health Organization European Standards for Drinking Water⁴ recommends a concentration of PAH not exceeding 0.2 µg/l.

Andelman and Suess¹ made calculations based on four samples of drinking water to determine an annual human consumption of PAH to be about 6, 9, 22, and 70 µg for the populations served by these water supplies.

Drinking water is not the only source of PAH for humans. They are also found in smoked and roasted foods, in vegetables, and plant fats and oils, as well as being breathed in from the atmosphere. Borneff⁷ warns that human exposure to PAH from all these sources could result in a yearly uptake of carcinogens at levels which cause carcinoma in animal experiments. The amount of carcinogenic PAH consumed from drinking water is probably not dangerous. Never-

theless, in combination with other sources it cannot be excluded as a possible hazard, especially since animal experiments indicate that repeated exposure to carcinogens is more effective in producing cancer than an equivalent single dose.

CONCLUSIONS

The following conclusions can be drawn from reviewing the literature:

1. The significance of traces of PAH in drinking water to human health is not yet known.

2. BP and other PAH are very insoluble in water but can be solubilized by detergents, oils, and other organic compounds. In most cases, however, the concentrations of detergents are not high enough to affect PAH solubilities.

3. BP can be very stable in water and is one of the most carcinogenic of the PAH.

4. With the present emphasis on water pollution control, attention should be given to reducing the amounts of carcinogenic PAH discharged to surface waters.

5. Concentrations of PAH in uncontaminated ground waters generally do not exceed 0.05 µg/l. Higher concentrations are found in surface and marine waters. Efforts should be made to determine the sources of the contamination of waters, especially when they may serve as sources of drinking water.

6. Mechanical separation processes, biological treatment, UV irradiation, and chlorination are not successful in removing enough carcinogenic PAH from drinking water. Ozonation and ClO₂ appear to be the most promising methods of effectively destroying PAH in drinking water.

7. The generally higher concentrations of PAH in polluted waters suggest that these compounds may have utility as indicators of pollution.

8. It would be prudent to establish MPD and MPC for PAH in drinking water.

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